PATENT SPECIFICATION

(11) **1 551 483**

551 483

20

(21) Application No. 1473/77 (22) Filed 14 Jan. 1977

(44) Complete Specification published 30 Aug. 1979

(51) INT CL² C08L 83/04, 23/18, 23/28 (C08L 23/18, 83/04) (C08L 23/28, 23/04)

(52) Index at acceptance

C3T 6A 6F2 6G2 6K2B 6K3 6K5 6K6 C3M 103 117 131 131C 132 132C 139 142 143 170 XA



(54) POLYMER COMPOSITIONS AND METHOD OF USING SAME TO PRODUCE THERMOSTABLE INSULATING MATERIALS

We, ISAAK YAKOVLEVICH PODDYBNY, of kvartira 47, bulvar Novatorov, 28, Leningrad, SERGEI VASILIEV-ICH AVERYANOV, of kvartira 18, prospekt Narodnogo Opolchenia, 181, Leningrad, LIDIA AKIVOVNA AVÉRYÁNOVA, of kvartira 18, prospekt Narodnogo Opolchenia, 181, Leningrad, MARK PEISAKHOVICH GRINBLAT, of kvartira 135, ulitsa Chernyshevskege, 9, Leningrad, RINAT RAUFÓ-VICH SAFIN, of kvartira 99, ulitza Zhdanova, 1, Kazan, ALEXANDR KHONONO-VICH BREGER of kvartira 131, ulitsa Polvarnava, 10, Moscow, VLADIMIR Polyarnaya, 10, Moscow, VLÁDIMIR ABRAMOVICH GOLDIN, of kvartira 147, ulitsa Markhlevskogo, 11, Moscow, ELENA ZINOVIEVNA BRANZBURG, of kvartira 29, korpus 2, ultisa Uralskaya, 6, Moscow, VIKTOR SEMENOVICH KONDRATOV-SKY, of kvartira 24, Pogonny proezd, 40, Moscow, ALBERT GUMEROVICH KABI-ROV, of kvartira 87, ulitsa Botanicheskaya, 13, Kazan, ALEXANDR TIMOFEEVICH

SANZHAROVSKY, of kvartira 186, korpus 3, Leninsky prospekt, 99, Moscow, VALERIA BORISOVNA SHTUKAREVA, of kvartira 85, Tishinskaya ploschad, 6, Moscow, JURY NIKOLAEVICH SVATIKOV, of kvartira 41, Buzheninovskaya ulitsa, 26/6,

Moscow and NADEZHDA MIKHAILOVNA
STEPANOVA, of kvartira 78, ulitsa Gastello,
41, Moscow, all Union of the Soviet Socialist
Republics, all citizens of the Union of Soviet
Socialist Republics, do hereby declare the invention, for which we pray that a patent may

vention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to polymer compositions and, more particularly, to polymer compositions based on polysiloxane and organic rubbers, as well as to methods of using such compositions to produce thermostable insulating materials.

5 Such compositions find extensive application in electrical engineering as insulating tapes, rubber-impregnated glass fabrics and varnished fabrics. They are also used as an electrical and thermal insulating material in the production and repair of electric equipment. Finally, such compositions are used to produce insulating and anticorrosion tapes to protect gas and oil pipelines.

It is only natural that stringent requirements are imposed on polymer compositions of this type, as well as on articles manufactured from them. Insulating tapes based on such compositions should possess the following properties:

1. The tape's adhesive coating is to ensure adhesion of overlapping layers of the tape over a period of 15 to 25 years despite the effects of moisture and elevated temperatures.

2. The adhesive layer is to impart certain dielectric properties to the insulating tape which must have an electric strength of no less than 3kV/mm and a volume resistivity of no less than 10¹⁰ ohm cm even after the effects of water.

Any method of producing thermostable insulating materials based on the polymer compositions in question largely determines the properties of articles manufactured from such compositions, since normally the materials and articles made therefrom are produced at the same time.

According to the invention, there is provided a polymer composition comprising 8 to 20 parts by weight of polyborsiloxane having a molecular weight of 2,000 to 5,000 and containing units:

$$\begin{bmatrix} \stackrel{\text{OH}}{\downarrow} & \stackrel{\text{H}_3C}{\downarrow} \\ B-O-Si \\ \downarrow & \\ H_3C \end{bmatrix} \text{ and } \begin{bmatrix} (CH_3)_2 SiO \end{bmatrix}$$

with a B:Si atom ratio of 1:4 to 5; 40 to 90 parts by weight of polysiloxane rubber; 10 to 60 parts by weight of an organic polymer

50

. .

60

65

70

75

55

60

65

70

10

20

which may be a polyolefine, a copolymer of an olefine and a diene, a halogenated polyolefine or a halogenated copolymer of an olefine and a diene, taken individually or in combination with one another; the total organic polymer and polysiloxane rubber being 100 parts by wt.; 2 to 10 parts by weight of a branched polyorganosiloxane of a molecular weight of 2,000 to 5,000 and the general formula:

$$\begin{bmatrix}
CH_3 \\
OSi \\
CH_3
\end{bmatrix}_{\alpha}
\begin{bmatrix}
CH_3 \\
OSi \\
CH=CH_2
\end{bmatrix}_{b}$$

and

wherein: $R = CH_3$ or C_0H_5 , a corresponds to 43 to 50 mole %. b corresponds to 0 to 2 mole %, c corresponds to 50 to 55 mole%; 10 to 25 parts by weight of silicon dioxide; and 5 to 10 parts by weight of a metal oxide thermal stabilizer.

The selected type of polyborsiloxane imparts the auto-adhesion capacity to the composition of this invention and articles produced from the composition.

Herein it is to be understood that the unit

$$\begin{bmatrix}
R \\
O-Si
\\
OH
\end{bmatrix}$$
and
$$\begin{bmatrix}
O \\
Si
\\
O
\end{bmatrix}$$

includes

An increase in the amount of polyborsiloxane in the composition above the maximum level reduces the hydrolytic stability of the composition, which makes subsequent treatment of the composition more difficult. A decrease in the amount of polyborsiloxane in the composition below the minimum level results in the composition and materials manufactured thereof not having sufficiently strong autoadhesion.

The polysiloxane rubber can be, for example, a dimethylsiloxane rubber containing

$$-\left\{ (CH_3)_2 SiO \right\}$$
 40

units, a vinylsiloxane rubber containing

$$\left\{ (CH_3)_2 SiO \right\} \text{ and } \left\{ \begin{array}{c} CH_3 \\ SiO \\ CH=CH_2 \end{array} \right\}$$

units, a methylphenylsiloxane rubber containing

$$\begin{array}{c|c} \hline (CH_3)_2 SiO \end{array} \quad \text{and} \quad \begin{array}{c|c} C_6H_5 \\ \hline SiO \\ \hline CH_3 \end{array} \quad 45$$

units, or a diphenylsiloxane rubber containing

$$+(CH_3)_2 SiO$$
and
 $+(C_6H_5)_2 SiO$
units.

The molecular weight of the selected polysiloxane rubbers is preferably between 300,000 and 900,000. Polysiloxane rubber of this molecular weight imparts strength, thermal stability and good dielectric properties to materials manufactured from the composition in accordance with the present invention. A decrease in the amount of rubber in the composition significantly affects all the abovementioned properties, while an increase in the rubber content impairs the adhesion and autoadhesion properties.

As stated above, the function of the organic polymer in the proposed composition can be performed by polyolefines, copolymers of olefines and dienes, halogenated polyolefines and halogenated copolymers of olefines and dienes. These components can be used individually or in any combination with one another. The organic polymers account for the composition's adhesive capacity at room temperature with regard to polymers and significantly improve the composition's adhesion to metals within the same temperature range.

The selected branched polyorganosiloxanes are characterized by their high viscosity at room temperature and reduced viscosity at a temperature of 70 to 80° C. For example, the viscosity of polydimethylphenylsiloxane is much higher at room temperature than that of low-molecular weight polydimethylsiloxane having a molecular weight of 5,000 to 40,000 and containing

10

25

45

50

$$-\left\{ (CH_p)_2 SiO \right\}$$

units, which is used according to the known

As a result, the composition of this invention and materials produced therefrom have a sufficiently high thermal stability. Furthermore, the compatibility of all the composition's components is remarkably pronounced. Without affecting the adhesive properties of the composition, branched polyorganosiloxane reduces the composition's viscosity with a reduction in the temperature, whereby subsequent treatment of the composition is greatly facilitated.

The viscosity of branched polyorganosiloxane depends on the type of radical of R which may be CH_3 or C_6H_5 . With $R = C_6H_5$, the viscosity of the polyorganosiloxane is higher than in the case of R = CH₃.

If the polyorganosiloxane contains vinyl units $(b \neq 0)$ it is more liable to further chemical transformations. Thus the type of branched polyorganosiloxane determines the viscosity and reactivity of the composition.

In the composition of this invention, silicon dioxide improves the mechanical properties of the composition. Pyrogenic types of silicon dioxide can be used, for example, different grades of "Aerosil" (a trade mark for pyro-gene silicon dioxide) with different specific surfaces. Precipitated silicon dioxide can also be used. The optimum content of silicon dioxide in the composition is 10 to 25 parts by weight.

The metal oxides are preferably of variable valency metals, for example, Fe₂O₃, Cr₂O₃ and TiO2. These oxides play the role of thermal stabilizers and are used in an amount of 5 to 10 parts by weight.

In the proposed polymer composition, the polyolefines can be, for example, polyisobutylene taken in an amount of 20 to 60 parts by weight. In order to facilitate processing, the molecular weight of polvisobutylene is preferably within the range of 5000 to 50,000. If the molecular weight of polyisobutylene is lower than 5,000, the composition adheres to the equipment employed in the process of its manufacture. A reduction in the polyisobutylene content below the minimum limit impairs the adhesive properties of materials produced from the composition of this invention. An increase in the polyisobutylene content above 60 parts by weight reduces the durability and thermal stability of the insulat-

ing materials.

The use of polyethylene, preferably with a molecular weight of 1,500 to 3,000, as the organic polymer results in suitably low viscosity levels of the polymer composition. This is of great importance when using polysiloxane rubbers with a molecular weight between 800,000 and 900,000. The optimum polyethylene content is 10 to 15 parts by weight, which results in a sufficiently high adhesive capacity of the composition, as well as a desired vis-cosity, and makes the polymer composition sufficiently workable. The above-mentioned molecular weight of polyethylene is selected because of the following considerations. First, it is quite difficult to synthesize polyethylene with a molecular weight below 1,500. On the other hand, polyethylene with a molecular weight of above 3,000 is a solid product which has to be ground to a required fineness in order to be introduced into the composition.

It is recommended that the polyolefine should be a copolymer of ethylene and propylene, preferably, with a molecular weight of 70,000 to 150,000, taken in an amount of 5 to 20 parts by weight and having a propylene content of 35 to 40 mole %. This copolymer improves the thermal stability of the composition and, accordingly, of materials produced from it. With the above-mentioned propylene content in the copolymer, the latter is a relatively soft type of rubber, which facilitates its introduction into the composition and improves the workability of the composition. It is expedient that this copolymer should be used in combination with other organic polymers.

It is advisable that the copolymer of olefine and diene should be a copolymer of isobutylene and isoprene, preferably, with a molecular weight of 3,000 to 60,000, taken in an amount 105 of 20 to 60 parts by weight and having an isoprene content of 0.6 to 3 mole %. It can also be a copolymer of ethylene, propylene and ethylidene norbornane, preferably, with a molecular weight of 70,000 to 150,000, taken in an amount of 5 to 20 parts by weight and having a propylene content of 35 to 40 mole % and an ethylidene norbornane content of 1 to 3 mole %.

On the one hand, the foregoing molecular 115 weights and ingredient ratios of the last three types of polymers result in satisfactory workability of the composition. On the other hand, they account for the required properties of the composition, such as adhesion, autoadhesion and thermal stability.

The halogenated polyolefines may be, for example, a brominated polyisobutylene taken in an amount of 20 to 60 parts by weight and having a molecular weight of 5,000 to 125 50,000; or a brominated copolymer of iso-

70

65

75

90

butylene and isoprene taken in an amount of 20 to 60 parts by weight and having an isoprene content of 0.6 to 3 mole % (the molecular weight of the latter copolymer is 3,000 to 60,000). The preferred bromine content for the halogenated polyolefine and halogenated copolymer is 2 to 3 percent by weight. These components make the composition fire-

The introduction into the polymer composition of N-bromosuccinimide also makes thermostable insulating materials produced fireproof. N-bromo-succinimide may be introduced in an amount of 2 to 6 parts by weight. This is an optimum amount at which the Npromosuccinimide is completely bonded with the organic polymer.

According to the invention, in a method for producing thermostable insulating materials the polymer composition of this invention is molded on a substrate to produce a blank which is exposed to ionizing radiation to reach an absorbed radiation dose of 6 to 35 megarads.

The substrate may be, for example, film of polyethylene, polyvinylchloride, or polyimide. It may also be paper impregnated, for example, with wax. Finally, it may be a fabric, such as organosilicon, capron, or a glass fabric.

The ionizing radiation source may be, for example, radioactive Coso, or high-speed elec-

Organic polymers, such as polyolefines, copolymers of olefines and dienes, and halogenated derivatives of these two types are known to degrade partially or completely after exposure to ionizing radiation. It is also known that polymer compositions of polysiloxane rubbers and polyborsiloxanes vulcanize under the action of ionizing radiation. It could be expected, therefore, that a composition comprising said components would degrade when exposed to ionizing radiation. However, in fact, the proposed polymer composition vulcanizes and allows the manufacture of thermostable insulating materials possessing all the required physico-mechanical properties. This is due to the fact that exposure of the proposed composition to ionizing radiation results in the formation of free radicals from the organic polymers (which partially degrade) and polysiloxane rubbers which interact with each other, whereby a common vulcanizing network is produced. The latter factor rules out further degradation of the organic polymers.

The co-vulcanization of the borsiloxane and siloxane components accounts for the autoadhesion of the insulating material. The organic polymers, which are grafted to siloxane units, account for the adhesion of the material to polymers and improve its adhesion

It is possible to manufacture articles of different configurations from thermostable insulating materials produced from the pro-

posed composition. The molding conditions are determined by the type of article to be manufactured. In some cases it is advisable that the polymer composition of this invention should be molded at a temperature of 60 to 100° C and a pressure of 20 to 100atm; this applies, for example, to the molding of plates of different shapes.

The foregoing temperature and pressure ranges are the optimum. Reducing the temperature and pressure below the minimum limit would impair the quality of materials (articles) being produced, whereas raising the temperature and pressure above the maximum level would make the polymer composition less fabricable (the composition adheres to the equipment).

In general, in order to raise the thermal stability of the materal, it is desirable that the polymer composition should be molded on a substrate which has already been exposed to ionizing radiation so that the adsorbed radiation dose amounts to 5 to 50 megarads. This is particularly advisable when the substrate is a film of polymers (polyethylene, polyvinylchloride) which cross-link when exposed to high-energy radiation. For instance, the thermal stability of non-irradiated polyethylene is 80°C. When irradiated with y-rays so that the absorbed radiation dose amounts to 25 megarads, the thermal stability of polyethylene reaches 110° C.

The optimum absorbed radiation dose of a blank which is molded on a substrate is 7 to 12 megarads.

At room temperature, thermostable insulating materials produced from the poposed polymer composition possess an adhesive capacity of about 3 to 6 kgf/cm² with regard to organic polymers, and 3 to 7 kgf/cm² with regard to metals. These matreials are autoadhesive at room temperatures.

Such materials possess satisfactory physicomechanical properties, including a sufficiently high thermal stability (130 to 150° C at prolonged exposures to elevated temperatures and 250° C at short exposures), cold resistance (-50° C), and ozone resistance. These properties remain intact even after the materials have been stored for 7 to 24 months at 20° C.

Such materials also possess satisfactory dielectric properties: their electric strength is 10 to 25 kV/mm, volume resistivity is 10¹² to 1013 ohm.cm, and permittivity is 3 to 3.5. These properties are not impaired by moisten-

For example, thermostable insulating tapes produced from such materials have a frost resistance factor of 0.1 to 1.0 at -50° C. As stated above, such tapes have an electric strength of 10 to 25 kV/mm, permittivity of 3 to 3.7, and volume resistivity of 10¹² to 10¹³ ohm.cm. Storage for 7 to 24 months does not affect these properties. The adhesive 130

70

85

95

100

110,

115

capacity of such tapes is preserved to a large extent (33 per-cent of the original value) even after 1,000 hours ageing at 110° C. Weight loss after ageing for 500 hours at 110° C

do not exceed 5 percent.

Due to the use of organic polymers in the proposed composition, polysiloxane rubber, silicon dioxide and oxides of variable valency metals are introduced in lesser quantities than in the case of similar compositions based on polysiloxane rubbers. This accounts for the above-mentioned combination of properties of thermostable insulating materials, which are superior to those of the known polymer compositions.

Insulating materials produced in accordance with the present invention can be used for cable connections and in the production of cable boxes intended for voltages of 1 to 35 kV; such materials are also used for anticorrosion insulation of gas and oil pipelines. For the latter purpose, such materials can be used in combination with conventional insulating materials of similar types.

The basic initial components for the production of the polymer composition of this invention, i.e. polysiloxane and organic poly-

mers, are readily available.

The process of producing thermostable insulating materials according to the invention is quite economical and makes it possible to manufacture articles of different types and configurations.

The proposed method for producing thermostable insulating materials is technologically simple and may be carried out as follows.

In order to prepare the composition, its components are mixed with the aid of rubber mixers or roller-type mixers at a temperature of 20 to 160° C. Preferably, the sequence of the production steps is this. First, there is added an organic polymer for example, polyisobutylene or a copolymer of isobutylene and isoprene or their halogenated derivatives, or polyisobutylene with N-bromosuccinimide. Then, to the organic polymer there are successively added polysiloxane rubber, polyborsiloxane, "Aerosil" (pyrogene silicon dioxide) with a low-molecular weight polyorganisiloxane, and a variable valency metal oxide.

If the organic polymer used in the process is liquid, for example, if it is low-molecular weight polyethylene, polysiloxane rubber is added first, whereafter there are added the organic polymer and polyborsiloxane. The remaining components are added as indicated above.

If use is made of a mixture of solid and liquid organic polymers, the components are introduced in the following order: the solid organic polymer, polysiloxane rubber, liquid organic polymer, and polyborsiloxane; the remaining components being added as indicated above.

.65

The result is a rubber mix which is successively passed through a refiner and a strainer in order to eliminate tramps.

The polymer composition thus produced is molded on a substrate, whereby there is produced a blank which is exposed to ionizing radiation to reach an absorbed radiation dose of 6 to 35 megarads, preferably, 7 to 12 megarads. The polymer composition may be molded at a temperature of 60 to 100°C and a pressure of 20 to 100 atm, for example, in a molding press. The molding conditions are determined by the type of article being manu-

In some cases it is preferable that the polymer composition should be molded on a substrate pre-irradiated to an absorbed radiation dose of 5 to 50 megarads.

For example, in order to manufacture insulating tape, the polymer composition is extruded or calendered onto a non-irradiated or irradiated substrate, for example, a film of polyethylene, in the form of a green band. The blank thus produced is exposed to ionizing radiation. The radiation source may be, for example, γ-radiation of Co₄₀, or highspeed electrons.

Rolls of finished tape are packed in polyethylene bags and shipped to consumers. Before use, the substrate is peeled off, so that only the rubber tape is used as insulation; the tape can also be used with the substrate. In both cases the insulating tape is wound around an article to be insulated which is kept for 6 to 48 hours at room temperature. 100 As a result, the tape layers effectively adhere to each other; the tape itself adheres to the article due to its high adhesion and autoadhesion capacity at room temperatures.

The advantages of the present invention 105 will be more readily understood from the following examples of preferred embodiments thereof.

Example 1. 45 parts by weight of methylvinylsiloxane 110 rubber having a molecular weight of 500,000 and a methylvinylsiloxane unit content of 0.07 mole %, 5 parts by weight of copolymer of ethylene and propylene with a propylene unit content of 35 mole %, 50 parts by weight of 115 polyisobutylene with a molecular weight of 19,000, 12 parts by weight of polyborsiloxane with a B:Si molar ratio of 1:5 and a moleculare weight of 2,000, 5 parts by weight of polydimethylphenylsiloxane with a molecular 120 weight of 2,000, 15 parts by weight of "Aerosil", and 5 parts by weight of Fe₂O₃ are mixed in a roller mixer at a temperature of 50 to 60° C. The rubber mix thus obtained is passed through a laboratory refiner with a 125 clearance between the rollers of less than 0.08 mm (this is done at the same temperature) and then through a strainer in order to separate tramps. The resultant polymer composition is listed in Table 1.

Example 2.

A polymer composition is prepared as in Example 1, but instead of the copolymer of ethylene and propylene the composition includes 5 parts by weight of the copolymer of ethylene, propylene and ethylidene norbornane having a molecular weight of 80,000, a propylene content of 35 mole % and an ethylidene norbornane content of 1.1 mole %. The resultant composition is listed in Table 1.

Example 3.

A polymer composition is prepared as in Example 1. The composition comprises 45 parts by weight of dimethylsiloxane rubber with a molecular weight of 400,000, 5 parts by weight of the copolymer of ethylene and propylene containing 35 mole percent of propylene units, 50 parts by weight of polyisobutylene with a molecular weight of 19,000, 10 parts by weight of polyborsiloxane with a B:Si molar ratio of 1:4 and a molecular weight of 5,000, 5 parts by weight of polydimethylphenylsiloxane having a molecular weight of 5,000, 15 parts by weight of "Aerosil", and 5 parts by weight of Fe₂O₃. The resultant composition is listed in Table 1.

Example 4.

A polymer composition is prepared as in Example 1. The composition comprises 43 parts by weight of dimethylsiloxane rubber having a molecular weight of 400,000, 7 parts by weight of the copolymer of ethylene and propylene having a molecular weight of 150,000 and a propylene unit content of 35 mole %, 50 parts by weight of polyisobutylene having a molecular weight of 19,000, 10 parts by weight of polyborsiloxane with a B:Si molar ratio of 1:4 and a molecular weight of 5,000, 5 parts by weight of polydimethylphenylsiloxane having a molecular weight of 5,000, 15 parts by weight of "Aerosil", and 5 parts by weight of Fe₂O₃. The resultant composition is listed in Table 1.

Example 5.

A polymer composition is prepared as in Example 3, but it contains 40 parts by weight of dimethylsiloxane rubber having a molecular weight of 450,000 and 10 parts by weight of the copolymer of ethylene and propylene of Example 4. The resultant composition is listed in Table 1.

Example 6.

A polymer composition is prepared as in Example 5, but it contains 12 parts by weight of polyborsiloxane of Example 4. The result55 ant composition is listed in Table 1.

Example 7.
A polymer composition is prepared as in

Example 1. The composition comprises 30 parts by weight of methylvinylsiloxane rubber having a molecular weight of 900,000 and a methylvinylsiloxane unit content of 0.07 mole %, 20 parts by weight of the copolymer of ethylene and propylene having a molecular weight of 70,000 and a propylene content of 35 mole %, 50 parts by weight of polyisobutylene having a molecular weight of 50,000, 8 parts by weight of polyborsiloxane with a B:Si molar ratio of 1:4 and a molecular weight of 5,000, 5 parts by weight of polydimethylmethylsiloxane having a molecular weight of 5,000, 12 parts by weight of "Aerosil", and 10 parts by weight of Fe₂O₃. The resultant composition is listed in Table 1.

Example 8.

40 parts by weight of dimethylsiloxane rubber having a molecular weight of 400,000, 10 parts by weight of the copolymer of ethylene, propylene and ethylidene norbornane having a molecular weight of 70,000 and a propylene content of 40 mole % and an ethylidene norbornane content of 3 mole %, 50 parts by weight of polyisobutylene having a molecular weight of 19,000, 9 parts by weight of polyborsiloxane having a B:Si molar ratio 1:4 and a molecular weight of 5,000, 3 parts by weight of polydimethylmethylvinylphenylsiloxane having a molecular weight of 3,000, a methylvinylsiloxane unit content of 2 mole % and a phenylsiloxane unit content of 43 mole %, 12 parts by weight of aerosil, and 5 parts by weight of Fe₂O₃ are mixed in a closed Banbury mixer during 45 minutes at a temperature of 85 to 160° C. The rubber mix is treated as in Example 1. The resultant polymer composition is listed in Table 1.

Examples 9 through 38.

Polymer compositions are prepared as in Example 1. The compositions are listed in Table 1.

According to Examples 9 through 19, 22, 23 and 31, use is made of the components of Example 1.

According to Example 20, use is made of a mixture of solid polyisobutylene with a molecular weight of 19,000 and liquid polyisobutylene having a molecular weight of 8,000, the weight ratio between the two being 4:1, respectively.

According to Example 21, use is made of polyisobutylene with a molecular weight of

According to Example 24, use is made of methylvinylsiloxane rubber having a molecular weight of 500,000 and a methylvinylsiloxane unit content of 1 mole %.

According to Example 27, use is made of brominated polyisobutylene with a molecular weight of 25,000 and a bromine content of 3 percent by weight.

•

70

75

0U

0.0

90

95

100

105

105

.

110

According to Example 33, use is made of the copolymer of isobutylene and isoprene having a molecular weight of 3,000 and an isoprene content of 0.6 mole %.

According to Example 34, use is made of the copolymer of Example 33, having a molecular weight of 60,000 and an isoprene

content of 3 mole %.

According to Example 35, use is made of polyisobutylene having a molecular weight of 19,000, and liquid polyethylene having a molecular weight of 1,500.

According to Example 36, use is made of liquid polyethylene with a molecular weight

of 3,000.

According to Example 37, use is made of the brominated copolymer of isobutylene and isoprene having a molecular weight of 60,000, an isoprene content of 0.6 mole % and a bromine content of 2 percent by weight.

According to Example 38, use is made of the brominated copolymer of isobutylene and isoprene with a molecular weight of 3,000, an isoprene content of 3 mole % and a bromine content of 3 percent by weight.

Example 39. The polymer composition of Example 1

is molded in the form of a cord of a round section with a diameter of 25 mm. This is done by passing the composition through an extruder. The cord is then passed through a profiling calender provided with a winding-up means. At the same time into the bite of the calender there is fed polyethylene film having a thickness of 60 mu. As a result, there is produced a blank in the form of a band which comprises the polymer composition molded on a substrate (the latter being the polyethylene film). The blank is 0.6 to 0.9 mm thick. The blank is wound up into rolls having a diameter of 10 to 12 cm and a width 8 to 9 cm, and is vulcanized through an exposure to γ-radiation of Co60. The absorbed dose is 7 megarads. The results of testing the thermostable insulating tape thus produced are listed in Table 2.

Example 40.

The polymer composition of Example 2 is molded as in Example 39 on a film of blackcontaining polyethylene (the black content amounts to 0.5 percent by weight). The film is 0.2 mm thick. The results of testing the tape are tabulated in Table 2.

Example 41.

The polymer composition of Example 3 is molded as in Example 39 in the form of a cord with a diameter of 7 mm, wherefrom there is produced a tape which is 25 mm wide. The blank is vulcanized till an absorbed radiation dose amounts to 8 megarads. The results of testing the tape are listed in Tables 2 and 3.

Examples 42 through 68. The polymer compositions of Examples 4. through 30 are molded as in Example 39.

According to all these examples, there is molded a cord with a diameter of 14 mm. The width of the insulating tape is 50 mm.

According to Example 43, the substrate is a pre-irradiated polyethylene film (the absorbed radiation dose is 5-megarads).

According to Example 45, the substrate polyvinylchloride film containing TiO₂. The film has a thickness of 0.15 mm.

According to Examples 46, 48, 58, 62 and 66, the substrate is polyvinylchloride film with a thickness of 0.1 mm.

According to Examples 47, 59, 64, 65 and 68, the substrate is varnished silicone fabric having a thickness of 0.15 mm; according to Example 61, the substrate is capron fabric with a thickness of 0.1 mm.

The absorbed radiation doses and the results of testing these tapes (without the substrates) are listed in Table 2. Additional information with regard to Examples 42, 46, 50, 51, 56, 57, 62 and 65 is contained in Table 3.

The insulating tape of Example 43 can be used with the substrate. In this case the tape's strength amounts to 33kgf/cm².

Example 69.

The polymer composition of Example 31 is placed in a press mold having inner dimensions of $120 \times 120 \times 1$ mm, whose bottom and cover are lined with tracing paper. The composition is then press-molded at a pressure of 100 atm and a temperature of 80° C during 15 minutes.

The blank thus obtained is exposed to ioniz- 100 ing radiation. The absorbed radiation dose and the results of testing are listed in Table 2.

Examples 70 through 74. The polymer composition of Examples 32 through 36 is molded as in Example 39.

According to Examples 70 and 73, the substrate is varnished silicone fabric having a thickness of 0.15 mm.

According to Example 71, the substrate is polyvinylchloride film with a thickness of 110 0.1 mm. The molded blank is covered with polyethylene film and exposed to ionizing radiation.

According to Examples 72 and 74, the substrate is polyethylene film having a thickness 115 of 60 mu.

The absorbed radiation doses and the results of testing the tapes thus produced (without the substrates) are listed in Table 2. Additional data with regard to testing the 120 tape of Example 74 are listed in Table 3.

The testing data with regard to Example 71 are related to an insulating tape with a substrate of polyvinylchloride.

COMPONENTS OF POLYMER COMPOSITION.

				-	Copolymers			_		
Serial No.	Polyiso- butylene	Copolymer of isobutylene and isoprene	Poly- ethylene	Brominated polyiso- butylene	Brominated copolymer of isobutylene and isoprene	Copolymer of ethylene and propylene	Copolymer of ethylene, propylene, ethylidene norbornane	N-bromo- succinimide	Dimethyl- siloxane rubber	Methyl- vinyl- siloxane rubber
1	2	3	4	5	. 6	7	8	9	10	11
1	50 .	. \ -		-	_	5	_	-	-	45
2	50)_	-	·· –	_	-	5	-	. –	45
3	50		_	-	_	5	-	-	45	_
4	50	_		_	_	7	-	-	43	-
5	- 50	. -	_	_		10	_	_	40	-
6	50	-	_		-	10	_	-	40	_
7	50	_	_	_	-	20 .	-	-	-	30
8	50	_	_	_	_	_	10		40	_
9	50	_		_	_	15	_	_	_	35.
10	50	_	_	_	. <u>-</u>	20	_	_	-	30
11	40	_	_	_	_	10	_	-	_	50
12	40		_	_	• _	10	_	-	_	50
13	40	. <u>.</u>	_		_	10	_	_		. 50
14	40	_	_	_	_	10	-	_	-	50
15	30	_	_		_	20	_	_	· _	50
16	50	_	_	_	_	_	_	_	-	50
17	50	_	_	_	-	_	_	<u>.</u>	_	50
18	50		_	_	_	_	 -	_	_	. 50
19	50	_	_	_	· _	_	_	_	_	50
20	50	<u>-</u>	_		_	_	_	<u> </u>	_	50
21	50	_	_	_	-	_	_	_		50
22	50	_	_		_	_	_	_	_	50
23	50	_	_	_	_	_	_	_		. 50
	50	_	_	_	_	_	_	6	_	50
24	50	-		_			-	2	_	. 50
25		-		_	_	_		_	_	50
26	50	_	_	50				_		50
27	-	-	-	-	_		_	_	_	40
28	60	_	10			_	_	_	_	40
29	50	_	15	_		_	_	_		40
30	·45	-		-		_		_	_	60
31	40	-	-	_	-	-		_	_	50
32	40	_	10	-	_	_	-	_	_	51
33	-	40	10	·• .·	-	_	-	, -	_	61
34		40	-	-	-	-		-	_	70
35	20	-	10		-	-		. –	-	
36	-	-	10	-	-	-	-	-	-	91
37	-	-	-	_	50 40	-	-	-	_	5

PARTS BY WEIGHT

	В	ranched polyorganos	iloxane				٠.	
Polybor- siloxane	Poly- dimethyl- phenyl- siloxane	Poly- dimethyl- methyl- siloxane	Poly- dimethyl- methyl- vinyl- phenylsi loxane	Aerosil	Precipitated SiO,	Fe,0,	TiO,	Cr ₂ O,
12	13	14	15	16	17	18	19	20
12	5	_		15	, -	5	<u> -</u>	-
12,	5	-	-	15.	-	5	- :	, -
10	· 5	-	. -	15	- :	5	-	-,
10	5		·	15	· -	5	_	· ·
10	· s	_	-	15	-	5	-	· –
12	5	-	-	15	-	5	-	- '
8	_	5	<u>-</u> ·	12	-	10	-	. -
9.	-	_	3	12	<u> </u>	5	- .	-
14	5	_		15	-	10	-	-
12	. 2	_	-	12	-	10		-
12	5	-	<u>.</u>	15	-	10		· -
9	5		_	15	-	5	-	
9	5	_	-	15	-	5	3	_
10	5	· ·-	-	15	-	10	. ""	
10	· 5	-		15	_	5	, -	_
12	· 5	-	- ·	15	· _	10	-	· -
. 12	5 -	-	_	15	-	5	-	
10	10	-	-	15	<u>,-</u>	5	. -	_
10	5	-	÷	15	-	, 5	. - .	-
10	5	-	-	15	·	, 5	<u>-</u>	
10	5	_	<u>-</u> .	15	-	5	-	-
9	5	-	- ·	15	· <u>-</u> ·	5	3.	- ,
8 -	5	-	-	15	-	5	-	-
8	1	-	-	18	-	5	· -	
8	1	_	-	18	-	5	-	
. 8	5	_	-	-	15	5	-	-
8	5	_	-	5	7.5	5	-	· -
8	5	-	-	18	· <u>-</u>	10	-	-
8	2	-	_	18	_	10		
8	1	-	, 	10	-	10	- '	-
. 8	2	_	-	10	-	5	· -	
8	3	, -	· -	10	-	10		- ,
20	3	-	- .	10	-	10	-	-
20	3	-	. –	15	-	5	.	-
8	5	=	-	-	20	10	_	-
8	5	-	- '	25	-	10	- ,	
8	2	= ;	- ·	10	-	10	-	-
8	1	-	-	10	-	-	-	- 5

	Œ		
	at		į
	8		3
	岩		
	an		
	EX		•
	.덤		
	88		
	ष्ठ		
	Plo		
	Έ.		1
	are		3
26	38		7
pu	ğ	ë	7
Š	ä	att	į
S 7	37	Š	4
ple	3	ಕ	•
cam	Ę	ដ្ឋ	F
斑	Exa	esst	į
	<u></u>	퉙.	2
		_	
,	Su	~	90
1	itions	and a	Paring
	positions	C and 8	of tracing
	ompositions	00°C and a	is of tracing
	r compositions	f 100°C and a	te is of tracing
	ymer compositions	e of 100°C and a	Strate is of tracing
	polymer compositions	ture of 100°C and a	substrate is of tracing
٠	he polymer compositions	verature of 100° C and a	he substrate is of traving
	The polymer compositions of Examples 37 and 38 are molded as in Example 69 at a	temperature of 100° C and a pressure of 20 atm.	The substrate is of tracing paper. The showted materials

			Swelling,	water	12	1.6	2.2	1.3	2.2	1.1	٠.			.			1.3	2.6	1
·			Equilibrium Swelling,	gasoline	11	249	244	291	327	230									
results are			kgf/cm²	to steel	10	7.1	5.9	6.9	5.0	0.9	7.0	6.0	5.5		5.8	8.9	5.4	5.0	5.4
imperature of 100° C and a pressure of 20 atm. The substrate is of tracing paper. The absorbed radiation doses and testing results are sted in Table 3.			Adhesion, kgf/cm²	to poly- ethylene	6	4.1	3.8	5.7	4.0	4.6	5.7	5.1	4.4		4.5	8.	3.6	3.6	4.1
adiation dos			ာ့	1	∞	53	28	92	89	48	09	15	70	. 99	32	16	52	.70	56
of 20' atm.	TABLE 2	ristics	g during 48 hours at 150°C	1	7	405	360	520	470	455	350	275	0.029	450	200	740	009	580	720
a pressure o	Ţ	al Characte	After Ageing during	Ъ	. 9	28	50	14	14	14	14	60	œ	6	4	15	14	11	13
100° C and is of tracing s.		Physico-Mechanical Characteristics		1	5	44	64	64	58	76	84	220	99	108	96	. 57	22	47	52
temperature of 100° C and a pressure of 20 atm. The substrate is of tracing paper. The absorbed listed in Table 3.		Physic	Initial at 20°C	١	4	850	845	086	026	1030	1170	1020	096	1160	1070	930	840	780	1100
temp T liste			II	Ь	3	11	14	12	. 15	14	7	9	10	12	ν	19	17	15	12
		. 1	Radiation	dose, - megarads	2	7	7	∞	∞	∞	7.5	∞	8.9	7	7	7	7.2	7.2	8.3
				Serial No.	. 1	39	40	41	42	43 ·	44	45	46	47	48	49	50	. 51	52

																• .			Ī
	12				2.6				2.2	2.4		· .					· ·		
	11			·	294								-						
	10	5.5	6.5	5.9	6.2	5.6		٠.	6.2	5.1	5.1	5.2	:		6.1	5.2			
	6	4.2	4.7	5.2	4.2	3.8	4.2	4.6	4.0	3.4	4.0	3.9	4.0	3.6	5.5	3.8	3.5	2.4	
(pənu	∞	27	44	12	40	55	11	40	48	14			28	18	99	100			
LE 2 (Continued)	7	089	570	200	575	575	285	069	390	260			595	355	210	350			
TABLE	9	12	15	24	17	20	20	23	15	22			22	20	9	v			
	5	56	26	46	62	. 21	46	09	40	30	28	64	24	32	89	96	40	12	
	4	810	825	780	1000	845	940	1240	790	750	755	1560	160	945	089	1230	1120	069	
	3	14	19	16	16	. 16	13	10	17	15	16	11	7	13	ý. 9	7	15	6	
	2	53 8.3 14 810	œ	∞	8.3	∞		10.1	7.2	œ	8.6	7.3	∞	7.4	3.5	8.5	10.2	11.2	
	-	53	54	55	95	57	. 28	59	09	61	62	63	64	65	99	. 19	89	69	

	12								
							•		
	11			,		378		204	
)	10			3.7					
-	6	3.1	3.6	2.7	1.5	6.0	2.3	3.1	
ned)	80			09	13	14			
TABLE 2 (Continued)	۲			630	310	330			٠.
TABLE	. 9				14	27		÷	
	5	12	'n	106	4	29	17	∞	
	. 4	855	200	1560	355	505	730	530	
	3	8	20	6	. 11	33	11	10	
	2	12	7	٠.	11.7	6	12.5	12.5	
	1	70	71	72	73	74	7.5	9/	

P is tensile strength, kgf/cm²

L is breaking elongation

¹ is permanent elongation, %

								771,7	-						
		n, kgf/cm²	to aluminium	7.2		7.0			• .	6.7					
		Adhesion	to copper	7.0		9.9	٠			6.5	6.3			.*	-
			-		10	41	19	88	38	99	25	18			
		250°C 1 hour	٦		75	245	105	260	70	460	210	70		•	
		1	d.		28	9	∞	vo.	9	\$	4	8			
		·	_	18	12	82	20		22	70		14	22	9	4
	ging	200°C 1 day	. L	95	7.5	330	145		140	225		110	120	165	240
щ Э	fter A		Ь	29	23	12	38		27	21		16	13	16	27
TABLE	stics A		-		12	30	32		٠.	4				e .	
·	Physical-Mechanical Characteristics After Aging	180°C 2 days	L		95	220	220			310					
	al Ch		Р	:	38	23	27			15					
	chanic	ø	120 days 15-20 days 2 days 1 day 1 hour Adhesion, P L 1 P L 1 TC copper	26	32	24	13	20	∞	•		.7	4.		
	ical-Me		L	185	130	235	250	65	235	190			20	06	
	Phys	15-	ď	19	707	14	25	13	27	32			40	35	
			1			•			. 9	9,				7	
		120°C 120 days	L					•	150	145				1.10	
			۵.				•		25	24				25	
,		-	Serial No.	41	. 42	46	47	·8	20	51	56	57	62	99	74

P, L and 1 are as in Table 2.

MARKS & CLERK, Chartered Patent Agents, 57-60 Lincolns Inn Fields, London, WC2A 3LS. Agents for the Applicants.

Printed for Her Majesty's Stationery Office by the Courier Press, Leamington Spa, 1979. Published by the Patent Office, 25 Southampton Buildings, London, WC2A 1AY, from which copies may be obtained.